

LONG-TERM RADIOACTIVE WASTE FROM FUSION REACTORS: PART II

Steve FETTER,¹ E.T. CHENG² and F.M. MANN³

¹School of Public Affairs, University of Maryland, College Park, MD 20742, USA

²GA Technologies, 10955 John Jay Hopkins Drive, San Diego, CA 92138, USA

³Hanford Engineering Development Laboratory, P.O. Box 1970, Richland, WA 99352

Abstract

In Part I we calculated 10 CFR 61 "Class-C" specific activity limits for all long-lived radionuclides with atomic number less than 88 (Ra). These calculations were based on the whole-body dose. We also estimated the production of these radionuclides from all naturally occurring elements with atomic numbers less than 84 (Po) in the first wall of a typical fusion reactor, and thereby derived concentration limits for these elements in first-wall materials, if the first wall is to be suitable for Class-C disposal. In Part II we use the "effective dose equivalent" (EDE), which is a much better indication of the risk from radiation exposure than the whole-body dose, to calculate specific activity limits for all long-lived radionuclides up to Cm-248. In addition, we have estimated the production of long-lived actinides and fission products from possible thorium and uranium impurities in first-wall structures. This completes our study of long-lived radionuclides that are produced from all elements that occur in the earth's crust at average concentrations greater than one part per trillion.

1. Introduction

In Part I, we noted that the potential of fusion to have lower radiological hazards than fission could be crucial to fusion's ultimate success as a commercial energy source [1]. In particular, a qualitative advantage over fission might be achieved if fusion reactors did not produce any high-level radioactive waste. To discover what restrictions this criteria would impose on the composition of fusion-reactor blanket materials, we first calculated the specific activity limits (SALs) for near-surface disposal of long-lived activation products, and then estimated the production of these radionuclides from many naturally occurring elements in the first wall of a typical reactor. The SALs were based on the whole-body dose to an inadvertent intruder into a waste disposal site, calculated using a model similar to that used by the U.S. Nuclear Regulatory Commission (NRC) in formulating its regulations of low-level radioactive waste disposal, known as "10 CFR 61" [2]. It should be noted that 10 CFR 61 regulations are directly relevant only for radioactive wastes produced in the United States.

The whole-body dose may not be the best indicator of risk from radiation exposure, however, because many radionuclides irradiate the body in a highly non-uniform way. In Part II, we use the "effective dose equivalent" rather than the whole-body dose to estimate specific activity limits. In addition, we have extended our calculations of SALs to include virtually all long-lived radionuclides, and we have applied these limits to the production of long-lived radionuclides from virtually all naturally occurring elements, including thorium and uranium.

2. 10 CFR 61 Specific Activity Limits

2.1. The 10 CFR 61 regulations

The guiding philosophy behind 10 CFR 61 is that no member of the public, at any time in the future, should be exposed to an unacceptable risk from accidental exposure to radioactive waste. Of the various exposure scenarios that were considered in drafting 10 CFR 61, the so-called "intruder" scenario produced the highest dose to individuals. The intruder scenario begins with the construction of a house on the waste-disposal site after the period of institutional control ends. The period of institutional control, which corresponds to the time period that governments can be expected to prevent access to the site, is assumed to be 100 years. Construction workers are exposed to direct gamma radiation from the waste and inhale waste particles while digging the foundation. If the waste is still stable—that is, recognizable to

the workers as radioactive waste—then construction is assumed to stop after six hours. Class C waste is assumed to be stable for 500 years.

If the waste is not stable, the workers do not realize that they are in a waste-disposal site and construction continues for 500 hours. The completed house is occupied, and the inhabitants inhale suspended waste particles and are exposed to direct gamma radiation from the waste. In addition, they are assumed to grow half of all their food—vegetables, meat, and milk—on the waste site. The inhabitants therefore ingest radionuclides deposited on the leaves of plants and absorbed through their roots, either directly in the case of vegetables, or indirectly through the meat and milk of cows in the case of grass. 10 CFR 61 limits the specific activity of radionuclides so that the 50-year whole-body dose commitment ("intruder dose") to workers from construction activity or the 50-year dose commitment to inhabitants from exposure during the first year does not exceed 0.5 rem (5 mSv), which is currently the maximum permissible dose per year for members of the public in the United States. The dose to any single organ was limited to 1.5 rem (15 mSv).

2.2. Effective dose equivalent

At dose rates this low, the risk from radiation exposure is due entirely to stochastic effects (e.g., cancer). If, as is usually assumed for regulatory purposes, the probability of developing a cancer is directly proportional to the radiation dose, then the risk of developing a fatal cancer in a particular organ is equal to the dose absorbed by that organ (rem) multiplied by the stochastic risk factor for that organ (fatal cancers/rem). Summing the probability of developing a fatal cancer in each organ over all organs gives the probability of a latent cancer fatality from a given exposure.

The effective dose equivalent (EDE) is an attempt to summarize this complicated situation in a single number. The EDE is the weighted average of the dose to certain organs, with the weights determined by the stochastic risk factors for the generation of fatal cancers associated with the respective organs as determined by the International Commission on Radiological Protection [3]. The weights are given in Table 1. The organ labeled "remainder" refers to the five organs, other than the skin or those listed above, receiving the highest doses; each of these is assigned a weight of 0.06. Thus, the EDE is approximately proportional to the probability of a latent cancer fatality—there is no need to consider the dose to each organ separately. Values for the 50-year EDEs for this study were calculated using a computer program developed by Fetter [4].

Table 1	
Weighting factors recommended by the ICRP for stochastic risks	
Organ or tissue	Weight
Gonads	0.25
Breast	0.15
Red bone marrow	0.12
Lung	0.12
Thyroid	0.03
Bone surfaces	0.03
Remainder	0.30

2.3. Specific activity limits

10 CFR 61 gives SALs for only a dozen radionuclides, and many of these are fission products or transuranics that are of little interest to fusion. We have developed a modified version of the NRC's intruder model to calculate limits for other long-lived radionuclides. This was an attempt to complement 10 CFR 61, not to replicate it, and there are several differences between the NRC model and the model used

here. First, several errors in the original NRC calculations were corrected; these corrections, which in most cases do not significantly affect the results, are described in Ref. 11. Second, the calculations done here are somewhat more detailed: plants were divided into two categories (produce and forage), more accurate transfer coefficients were used, and the shielding provided by overlaying soil was calculated for each radionuclide. Third, as described above, the 50-year EDE is limited to 0.5 rem (5 mSv). In view of the huge uncertainties in many variables of the model, we do not necessarily believe that our estimates are more accurate those given in 10 CFR 61, but we do think that the relative hazard of different radionuclides is better represented by our SALs.

Table 2 gives the SALs (based on the EDE) for Class C disposal of all known radionuclides with half-lives greater than 5 years and less than 10^{12} years and with atomic numbers up to 96 (Cm) [5]. Radionuclides with half-lives less than 5 years are not limited by 10 CFR 61 since it is assumed that wastes can be isolated from the public for at least 100 years. Radionuclides with half-lives greater than 10^{12} also pose no hazard because their rate of decay is extremely small. Thus, Table 2 represents all of the potentially hazardous long-lived radionuclides that could be produced in quantities from naturally occurring elements.

Also listed in Table 2 are the SALs given in 10 CFR 61. When comparing these SALs to our values, one should bear in mind that the 10 CFR 61 SALs were based on a worst-case waste form; that is, they generally do not take proper account of the fact that, for many fusion wastes, the radionuclides would be embedded in metal. 10 CFR 61 allows a factor of ten reduction in the SAL if the radionuclide is embedded in activated metal (this is included in all cases), but this factor of ten has basis in the model only when the intruder dose is dominated by external radiation. If the intruder dose is dominated by internal radiation (i.e., if it does not emit strong gamma rays), then the reduction due to the added stability of metal waste can be very much greater. The NRC decided not to develop separate regulations for activated metal waste because they currently represent a small fraction of the total waste stream. Since fusion would generate a significant amount of activated waste, the NRC position must be reevaluated.

As anticipated, the use of the whole-body dose in Part I did not lead to great errors in the SALs compared to the limits based on the EDE given here. Although the SALs for several radionuclides changed by modest amounts, the SALs for only two radionuclides changed by more than a factor of two: Tc-99 (whole-body dose gave a limit three times higher than the EDE), and Sn-121m (whole-body dose gave a limit seven times lower than the EDE).

3. Long-term radioactivity in fusion materials

In Part I we described our method of estimating the production of long-lived radionuclides induced in fusion reactor materials [1]. Briefly, the neutron fluxes used were those typical of the first wall of a lithium-cooled blanket with vanadium-alloy structure. The first wall is 1 cm thick and is composed of 40% vanadium alloy, 37.5% lithium, and 22.5% void by volume. Behind the first wall is a 1 cm gap, a 30 cm thick blanket, a 25 cm thick reflector, and a 20 cm thick shield. The blanket and reflector are composed of 20% vanadium alloy and 80% lithium by volume; the shield is 90% vanadium alloy and 10% lithium. The Li-6 content in the lithium coolant was varied from 2% to 75% to explore the effect on the neutron spectrum. The total neutron fluence was 20 MW-y/m².

Combining the estimates of radionuclide production with the specific activity limits in Table 2 yields concentration limits for elements in the first-wall material. Table 3 gives concentration limits for every element that is present in the earth's crust at concentrations greater than one part per trillion [10].

As indicated by Table 3, several potential first-wall materials may not be compatible with the goal of producing only low-level waste. In particular, any first-wall material that contains more than 0.1% aluminum or 1 ppm molybdenum may not be acceptable. (As indicated by the table, some of these limits depend sensitively on Li-6 enrichment, and therefore on reactor design.) Silicon carbide and other silicon-based materials may also not be acceptable in the first wall, since the limit for silicon is a factor of ten lower if it is not contained within a metal (i.e., 3% instead of 30%).

Moreover, careful attention should be given to some elements as impurities in reactor materials. The limits for niobium, molybdenum, gadolinium, terbium, and holmium are of the same order as or less than their natural abundance in the earth's crust. Silver, erbium, and thorium could also present problems. For certain metals, cadmium, europium, dysprosium, hafnium, osmium, bismuth, and uranium impurities may have to be controlled. The fact that many of these limits are on the order of 1 ppm underscores the importance of accurate assays of materials under consideration.

It should be emphasized that these calculations are based on the neutron flux in a vanadium first wall. They may not be appropriate for determining the permissible concentrations of elements in blanket materials, especially if the neutron reactions that produce the hazardous radionuclides have energy thresholds of a few MeV or higher, as would be case for those radionuclides produced predominately by (n,2n) or (n,3n) reactions. In the case of aluminum, for example, the hazardous radionuclide Al-26 is produced by (n,2n) reactions with the sole isotope of aluminum, Al-27. Thus, aluminum that is located deep enough in the blanket to minimize the fast neutron flux may be suitable for disposal as Class C waste.

4. Dilution and averaging

Some fusion researchers unfamiliar with the U.S. debate on low-level radioactive waste disposal have assumed that the Class C disposal criteria could be met for any material, no matter how radioactive, simply by dilution. It has been suggested, for example, that filling the void spaces in the blanket with grout or chopping up the first wall and mixing it with a sufficient quantity of concrete would be acceptable solutions when the first wall is too radioactive to be disposed of normally.

Dilution is unlikely to be acceptable as a routine solution for a number of reasons. First, "dilution as the solution to pollution" has been rejected; the trend has for some time been toward volume minimization. During congressional hearings on 10 CFR 61, legislators and lobbyists expressed concern that high-level waste could be diluted and disposed of as low-level waste. The NRC offered repeated assurances that this would not be the case. Second, space at low-level waste disposal sites is likely to be limited for some time, and the NRC will not look kindly on requests to greatly increase the volume of a waste stream. Third, dilution has generally been allowed in the past only when it was necessary to improve the stability or handling of the waste. It is unlikely, however, that the stability of fusion-reactor blankets can be "improved" by diluting them. On the other hand, "averaging," or mixing less-radioactive wastes with more-radioactive wastes, might be allowed if the mixing process does not decrease the stability of the waste or create significant occupational risks. If blanket modules are most easily and safely handled and disposed of in one piece, then the NRC might allow averaging the activity over the total blanket volume. As noted above, this could significantly increase the allowable concentrations of certain elements if the hazardous radionuclides are produced by threshold reactions.

5. Conclusions

We have evaluated, using a model similar to that used to formulate the 10 CFR 61 regulations, the specific activity limits for Class C, shallow land burial of long-lived radionuclides that could be induced in fusion reactor materials. These limits were based on the effective dose equivalent rather than the whole-body dose. The specific activity limits were used to determine the concentration limits, for materials located in the first-wall region, of all naturally occurring elements. We find that the use of aluminum and molybdenum is severely restricted in first-wall materials, and that niobium, gadolinium, terbium, and holmium impurities will have to be strictly limited.

This completes our study of long-lived radioactivity in the first wall of a generic fusion reactor. To evaluate fusion wastes more accurately, specific reactor designs must be considered.

References

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Table 2

Specific activity limits for class "C" disposal of activated metal for all radionuclides with $5 \text{ y} < t_{1/2} < 10^{12} \text{ y}^a$

Radio-nuclide	Halflife	SAL (Ci/m ³) ^b	Other values ^d
H-3	12.3 y	TMSA ^c	TMSA (10 CFR 61)
Be-10	1.6 My	5.E+03	7,000 [7]; 3 [8]
C-14	5.7 ky	6.E+02 - 6.E+03	80 (10 CFR 61)
Al-26	720. ky	9.E-02	0.1 [6]
Si-32	104. y	6.E+02 - 4.E+03	600 [7]; 30 [8]
Cl-36	301. ky	1.E+01 - 1.E+02	3 [8]
Ar-39	269. y	2.E+04	2,000 [8]
Ar-42	33. y	2.E+04	0.8 [6]; 7,000 [8]
K-40	1.3 Gy	2.E+00	
Ca-41	103. ky	1.E+04 - 3.E+04	3 [8]
Ti-44	47. y	2.E+02	0.06 [6]; 300 [8]
Mn-53	3.7 My	TMSA	600 [7]; 30 [8]
Fe-60	100. ky	1.E-01	0.01 [6]; 0.1 [8]
Co-60	5.3 y	3.E+08	TMSA (10 CFR 61)
Ni-59	75. ky	9.E+02	220 (10 CFR 61)
Ni-63	100. y	7.E+05 - 7.E+06	7,000 (10 CFR 61)
Se-79	65. ky	5.E+01 - 5.E+02	3 [8]
Kr-81	210. ky	3.E+01	300 [8]
Kr-85	10.7 y	TMSA	
Rb-87	48. Gy	TMSA	
Sr-90	28.5 y	8.E+05 - 7.E+06	70,000 (10 CFR 61) ^e
Zr-93	1.5 My	TMSA	200 [7]; 10 [8]
Nb-91	680. y	2.E+02	
Nb-92	36. My	2.E-01	0.3 [6]
Nb-93m	13.6 My	TMSA	
Nb-94	20. ky	2.E-01	0.2 (10 CFR 61)
Mo-93	3.5 ky	4.E+03	30 [7,8]
Tc-97	2.6 My	4.E-01 - 4.E+00	
Tc-98	4.2 My	1.E-02 - 8.E-02	0.02 [6]
Tc-99	213. ky	6.E-02 - 6.E-01	30 (10 CFR 61) ^e
Pd-107	6.5 My	9.E+02 - TMSA	
Ag-108m	127. y	3.E+00	3 [6,8]
Cd-113m	13.7 y	TMSA	
Sn-121m	55. y	7.E+05	3,000 [8]
Sn-126	100. ky	1.E-01	0.01 [6]
I-129	15.7 My	2.E+00 - 1.E+01	0.8 (10 CFR 61) ^e
Cs-135	3.0 My	TMSA	8,400 (10 CFR 61) ^e ; 3 [8]
Cs-137	30.0 y	5.E+04	46,000 (10 CFR 61) ^e
Ba-133	10.5 y	2.E+08	55 [6]
La-137	60. ky	2.E+02	
La-138	106. Gy	TMSA	
Pm-145	17.7 y	TMSA	
Pm-146	5.5 y	TMSA	

Table 2 (cont.)

Radio-nuclide	Half-life	SAL (Ci/m ³) ^b	Other values ^d
Sm-146	103. My	TMSA	
Sm-147	106. Gy	TMSA	
Sm-151	90. y	5.E+07 - TMSA	3,000 [8]
Eu-150m	36. y	3.E+03	3,000 [8]
Eu-152	13.3 y	3.E+05	
Eu-154	8.8 y	5.E+06	
Gd-148	98. y	2.E+05 - 2.E+06	
Gd-150	1.8 My	2.E+03 - TMSA	
Tb-157	150. y	5.E+03	
Tb-158	150. y	4.E+00	5 [8]
Dy-154	10. My	1.E+03 - TMSA	
Ho-166m	1.2 ky	2.E-01	0.2 [8]
Lu-176	35.9 Gy	TMSA	
Hf-178m	31. y	9.E+03	0.25 [6]; 3,000 [8]
Hf-182	9. My	2.E-01	0.02 [6]
Re-186m	200. ky	2.E+01	10 [8]
Re-187	40. Gy	TMSA	
Os-194	6.0 y	TMSA	
Ir-192m	241. y	1.E+00	1 [8]
Pt-190	600. Gy	TMSA	
Pt-193	50. y	2.E+08	
Hg-194	520. y	5.E-01	
Pb-202	53. ky	6.E-01	0.07 [6]
Pb-205	19. My	TMSA	5 [7]; 3 [8]
Pb-210	22.3 y	3.E+07 - 3.E+08	
Bi-207	32.2 y	9.E+03	17,000 [6]
Bi-208	368. ky	8.E-02	0.1 [6,8]
Bi-210m	3.0 My	1.E+00	2 [6]; 0.5 [8]
Po-209	102. y	3.E+03	
Ra-226	1.6 ky	1.E-01 - 2.E-01	
Ra-228	5.8 y	3.E+07	
Ac-227	21.8 y	5.E+05 - 2.E+06	
Th-229	7.3 ky	2.E+00	
Th-230	75. ky	3.E-01	
Th-232	14.0 Gy	1.E-01	
Pa-231	62.8 ky	7.E-01	
U-232	68.9 y	3.E+01	
U-233	159.2 ky	2.E+01	
U-234	245. ky	9.E+01	
U-235	703.8 My	2.E+00	4 [9]
U-236	23.4 My	TMSA	
U-238	4.5 Gy	TMSA	TMSA [9]
Np-236	115. ky	1.E+00	6 (10 CFR 61) ^e
Np-237	2.1 My	1.E+00	6 (10 CFR 61) ^e ; 0.4 [9]

Table 2 (cont.)

Radio-nuclide	Half-life	SAL (Ci/m ³) ^b	Other values ^d
Pu-238	87.7 y	7.E+04 - 5.E+05	6 (10 CFR 61) ^e ; 70 [9]
Pu-239	24.1 ky	1.E+03 - 5.E+03	6 (10 CFR 61) ^e ; 1 [9]
Pu-240	6.6 ky	1.E+03 - 1.E+04	6 (10 CFR 61) ^e ; 1 [9]
Pu-241	14.4 y	2.E+03	200 (10 CFR 61) ^e ; 50 [9]
Pu-242	373.3 ky	1.E+03 - 1.E+04	6 (10 CFR 61) ^e ; 1 [9]
Pu-244	80.8 My	9.E-01	6 (10 CFR 61) ^e
Am-241	432.2 y	5.E+01	6 (10 CFR 61) ^e ; 1 [9]
Am-242m	141. y	3.E+02	
Am-243	7.4 ky	2.E+00	6 (10 CFR 61) ^e ; 0.7 [9]
Cm-243	28.5 y	6.E+02	6 (10 CFR 61) ^e ; 800 [9]
Cm-244	18.1 y	5.E+05 - 4.E+06	6 (10 CFR 61) ^e ; 400 [9]
Cm-245	8.5 ky	5.E+00	6 (10 CFR 61) ^e
Cm-246	4.8 ky	8.E+02 - 8.E+03	6 (10 CFR 61) ^e
Cm-248	340. ky	8.E+02 - 8.E+03	6 (10 CFR 61) ^e

^aExcept Cm-247, Bk-247, Cf-249, Cf-250, and Cf-251

^bSpecific Activity Limits (SALs) depend on waste form indices. The indices used here are appropriate for activated metal that may or may not be corroded, and are the same as those used in the environmental impact statement of 10 CFR 61 for non-fuel reactor components (lower limits) and high-activity industrial waste (higher limits). The limits published in 10 CFR 61 are for a worst-case waste form; these often differ substantially from the limits given here when the intruder dose is not dominated by direct gamma radiation.

^cTheoretical Maximum Specific Activity (i.e., the activity of 1 m³ of the pure radionuclide at normal density).

^dValues are for radionuclides contained in or permanently fixed to metal.

^eThe 10 CFR 61 limits for Sr-90, Tc-99, I-129, Cs-137, Pu-241, and alpha-emitting transuranics are multiplied by a factor of ten because they are assumed to be contained in activated metal.

Table 3

Concentration limits for all naturally occurring elements in a metal first wall

Z		Concentration Limit	Major Contributors		Z		Concentration Limit	Crustal Abundance (ppm)	Major Contributors	
1	H	—		d	41	Nb	0.4-9 ppm	20	Nb-94	a
2	He	—			42	Mo	0.2-2 ppm	1.5	Tc-99, Tc-98	c
3	Li	—			44	Ru	0.01-0.1%	0.001	Tc-99, Nb-94	c
4	Be	—			45	Rh	0.02-0.2%	0.001	Tc-99	c
5	B	—			46	Pd	20-30 ppm	0.01	Ag-108m	a
6	C	—			47	Ag	0.5-1 ppm	0.07	Ag-108m	a
7	N	3-40%	C-14	a,c	48	Cd	30 ppm	0.2	Ag-108m	
8	O	—			49	In	2%		Ag-108m	
9	F	—			50	Sn	10-30%		Sn-121m, Ag-108m	a
10	Ne	—			51	Sb	80-100%		Sn-121m	a
11	Na	—			52	Te	—			
12	Mg	—			53	I	—			
13	Al	0.1%	Al-26	e	54	Xe	—			
14	Si	30%	Al-26	e	55	Cs	—			
15	P	—			56	Ba	70-90%		Cs-137	b
16	S	50-100%	Si-32	a,c	57	La	20%		La-137	
17	Cl	20-100%	Cl-36, Si-32	a,c	58	Ce	80-100%		La-137	a
18	Ar	2%	Ar-39		59	Pr	—			
19	K	2-3%	Ar-39, Cl-36	c	60	Nd	—			
20	Ca	20%	Ar-39		62	Sm	7-10%		Eu-152, Eu-154, Eu-150m	a
21	Sc	—			63	Eu	200-300 ppm	1.2	Eu-150m, Eu152	b
22	Ti	—			64	Gd	4-20 ppm	5.4	Tb-158	a
23	V	—			65	Tb	0.2-0.3 ppm	0.9	Tb-158	b
24	Cr	—			66	Dy	200 ppm	3.0	Tb-158	
25	Mn	—			67	Ho	0.4-10 ppm	1.2	Ho-166m	a
26	Fe	—			68	Er	20-70 ppm	2.8	Ho-166m	a
27	Co	9-100%	Co-60	a	69	Tm	200-300 ppm	0.47	Ho-166m	b
28	Ni	10-15%	Fe-60, Ni-59	a	70	Yb	—			
29	Cu	30-100%	Fe-60, Ni-63	a,c	71	Lu	0.03-100%	0.5	Hf-178m	a
30	Zn	—			72	Hf	80-300 ppm	2	Hf-178m	a
31	Ga	—			73	Ta	6-8%		Hf-178m	a
32	Ge	—			74	W	15%		Hf-178m	
33	As	—			75	Re	5-1000 ppm	0.005	Ir-192m, Re-186m	a
34	Se	0.3-3%	Se-79	c	76	Os	0.1-10 ppm	0.0015	Ir-192m	a
35	Br	2-40%	Kr-81, Se-79	a,c	77	Ir	0.05-0.1 ppm	0.001	Ir-192m	a
36	Kr	7-8%	Kr-81	a,c	78	Pt	20-100 ppm	0.005	Ir-192m	a
37	Rb	—			79	Au	0.4-0.9%		Ir-192m	b
38	Sr	60-100%	Se-79, Kr-81	c	80	Hg	70-100%		Ir-192m	b
39	Y	—			81	Tl	—			
40	Zr	3-4%	Nb-94	a	82	Pb	10-20%		Bi-208	a
					83	Bi	20-30 ppm	0.17	Bi-208	b
					90	Th	100 ppm	9.6		
					92	U	1000 ppm	2.7		

^aLimit depends on Li-6 enrichment; limits are higher for 75% Li-6.

^bLimit depends on Li-6 enrichment; limits are higher for 2% Li-6.

^cLimit depends on waste-form indices; limits are higher for less corroded material.

^dLimit greater than 100%.

^eLimit is factor of ten lower if element is not in metal (e.g., LiAlO₂ or SiC).